Spin-atomic vibration interaction and spin-flip Hamiltonian of a single atomic spin

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Recently, magnetic properties for molecular magnets and atomic spins have been extensively studied toward the development of ultimate microscopic elements for mass-storage devices and quantum information devices [1-3]. In the field of data storage, quantum spin systems with bistable states, which contribute to 1 bit of information storage, are expected to be an ideal memory element. A typical energy producing the bistable states is a uniaxial anisotropy energy, $-|D|S_z^2$, with D being the uniaxial anisotropy constant. Materials with such an energy are Mn_{12} of S=10 [4] with $|D|$=0.06 meV and a single Fe atom on a CuN surface of S=2 with $|D|\simeq$1.55 meV [1]. In particular, this Fe atom may have the potential of a single atomic memory.

Regarding the spin system with $-|D|S_z^2$, it is known that the spin relaxation has a strong influence on the spin switching time (i.e., the writing time of data), and so on [3]. An origin of the spin relaxation is considered to be the spin-atomic vibration interaction V_{SA}, because the atomic vibration energy is usually in the range of 0.041 meV - 41 meV (10^{10} s^{-1} - 10^{13} s^{-1}) which is comparable to energy-level spacings of spin systems. To our knowledge, however, the concrete expression of V_{SA} has not been reported so far.

In this paper, we derived V_{SA} and the spin-flip Hamiltonian V_{SF} of a single atomic spin in the crystal field, using the perturbation theory for the spin-orbit (SO) interaction in which the difference of displacement between the nucleus and the electron, $\Delta\vec{r}$, is taken into account (see Fig. 1). For the case of Fe^{2+}, we investigated the presence or absence of V_{SA} and V_{SF} for any parameter sets. In addition, the magnitude of their coefficients was roughly estimated.

The perturbation energy for the SO interaction is obtained as, $V = V_A + V_{SA} + V_{SF}$, with

$$V_A = DS_z^2 + E(S_x^2 - S_y^2), \tag{1}$$

$$V_{SA} = \sum_{\mu,\nu=x,y,z} S_\mu \left(\Lambda^{(1)}_{\mu,\nu} a_\nu + \Lambda^{(2)}_{\mu,\nu} a^\dagger_\nu\right) + \sum_{\mu,\nu,\xi=x,y,z} S_\mu S_\nu \left(\Lambda^{(1)}_{\mu,\nu,\xi} a_\xi + \Lambda^{(2)}_{\mu,\nu,\xi} a^\dagger_\xi\right), \tag{2}$$

$$V_{SF} = \sum_{\mu,\nu=x,y,z} \Gamma_{\mu,\nu} S_\mu S_\nu. \tag{3}$$

Here, V_A is the so-called anisotropy spin Hamiltonian [5], where E is the biaxial anisotropy constant. The operator S_μ is the spin operator in the direction of μ, and $a^\dagger_\nu$ (a_ν) is the creation (annihilation) operator of the atomic vibration in the direction of ν. The coefficients $\Lambda^{(i)}_{\mu,\nu}$, $\Lambda^{(i)}_{\mu,\nu,\xi}$, and $\Gamma_{\mu,\nu}$ contain the matrix element of the orbital angular momentum, and so on.

We now focus on Fe^{2+} ($3d^6$) in a crystal field of the tetragonal symmetry. In this case we consider only one down-spin electron because the up-spin shell is filled. The abovementioned coefficients are therefore calculated by using the following orbital state:

$$|\phi_i\rangle = C_i \left(|d_i\rangle + \sum_{d_j(\neq d_i)} c_{d_j}^{(i)} |d_j\rangle + \sum_p c_p^{(i)} |p\rangle \right), \tag{4}$$

with $C_i{=}(1+\sum_{d_j(\neq d_i)} |c_{d_j}^{(i)}|^2 + \sum_p |c_p^{(i)}|^2)^{-1/2}$, $|c_{d_j}^{(i)}|^2 \ll 1$, and $|c_p^{(i)}|^2 \ll 1$, where the energy level for $|\phi_i\rangle$ is written as E_i. Here, $|d_i\rangle$ is the dominant d orbital, while $|d_j\rangle$ and $|p\rangle$ are the other d orbital and the p orbital in the atom, respectively. Owing to the d-d and d-p hybridizations in the atom, $|d_j\rangle$ and $|p\rangle$ are included in $|\phi_i\rangle$. The hybridizations originate from, for example, the mixing of atomic orbitals via the surrounding ions.

On the basis of expressions of the coefficients, we investigate the presence or absence of V_{SA} and V_{SF}, where $c_{d_j}^{(i)}{=}c_d$ and $c_p^{(i)}{=}c_p$ are set (see Table 1). The interaction V_{SA} exists for $\Delta\vec{r}\neq 0$ and $c_p\neq 0$, although it vanishes for $\Delta\vec{r}=0$. Namely, the d-p hybridizations as well as $\Delta\vec{r}\neq 0$ play an important role in the presence of V_{SA}. On the other hand, V_{SF} is present for $c_d\neq 0$ even when $\Delta\vec{r}=0$. The d-d hybridization is essential for the presence of V_{SF}.

When $|\Delta\vec{r}|/|\Delta\vec{r}_n|{=}0.5$, $c_d{=}c_p$, and $\sum_d |c_d|^2 + \sum_p |c_p|^2{=}0.2$ are set, where $\Delta\vec{r}_n$ is the displacement of the nucleus, we estimate the magnitude of the coefficients of V_{SA} and V_{SF} as follows: The largest coefficient of V_{SF} divided by $|D|$ is 0.2, while that of the second term of V_{SA} divided by $|D|$ is 0.1. Also, the largest coefficient of the first term of V_{SA} divided by $|\lambda|$ is less than 10^{-4}, where λ is the SO interaction constant.

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Figure and Table:

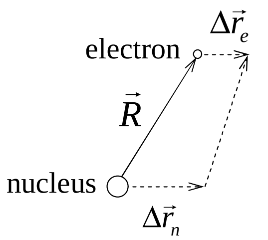

Fig. 1 : Positions and displacements of the nucleus (large circle) and the electron (small circle). The difference of displacement between the nucleus and the electron is given by $\Delta r{=}\Delta\vec{r}_e - \Delta\vec{r}_n$, where $\Delta\vec{r}_n$ is the displacement of the nucleus, and $\Delta\vec{r}_e$ is that of the electron. In addition, $\vec{R}$ is the position vector of the electron measured from the nucleus.

Tabel 1 : The presence or absence of V_{SA} and V_{SF} for each set of $\Delta\vec{r}$, c_d, c_p. The presence and absence are represented by ◯ and ×, respectively.

		V_{SA}	V_{SF}
$\Delta\vec{r}=0$	$c_d=0$, $c_p=0$	×	×
	$c_d=0$, $c_p\neq 0$	×	×
	$c_d\neq 0$, $c_p=0$	×	◯
	$c_d\neq 0$, $c_p\neq 0$	×	◯
$\Delta\vec{r}\neq 0$	$c_d=0$, $c_p=0$	×	×
	$c_d=0$, $c_p\neq 0$	◯	×
	$c_d\neq 0$, $c_p=0$	×	◯
	$c_d\neq 0$, $c_p\neq 0$	◯	◯